**GLYCOLYTIC RECYCLE OF PET FOR PRODUCTION OF HIGH PURITY BHET.
AN EXAMPLE OF CONCEPTUAL AND BASIC DESIGN
OF A SEMICONTINUOUS SOLID PROCESS**

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GLYCOLYTIC RECYCLE OF PET FOR PRODUCTION OF HIGH PURITY BHET.
AN EXAMPLE OF CONCEPTUAL AND BASIC DESIGN OF A SEMICONTINUOUS SOLIDS PROCESS

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Abstract - The design of an innovative glycolytic process for PET recovery is presented as an example of the approach to teaching process design at Politecnico di Torino. The process investigated is a typical polymer process which include solid handling steps, with part of the operations carried out batchwise.

Introduction

In the past, in the chemical engineering science, continuous processes were given much more attention than batch and semi-continuous processes; this is a consequence of the greater difficulty involved in design of batch processes and of the fact that the design procedure can not be easily generalised. As far as continuous operations are concerned, the processes that deal with fluids have been more deeply and frequently investigated under a fundamental viewpoint than those which handle particulate solid material.

In spite of the relevance that the above mentioned processes have currently in the chemical plants, as number of applications even though usually for small productions, relatively little attention is often given to the conceptual design of non-continuous and solid-handling processes in the chemical engineering courses at University. At Politecnico di Torino, process and equipment design is currently taught in two subsequent courses: in the first one the students are taught the hierarchical approach to conceptual design according to Douglas (1985); in the second one, a novel project is developed by the teaching staff together with a student team starting from the conceptual stage up to completing the basic engineering design, with the emission of the process P&ID, the design report and the operative manual. Either data from literature or patents or experimental data obtained in bench scale or pilot plant apparatus are utilised as input. In this paper an example of the work performed is presented, to stimulate discussion on such didactic approach.

An innovative glycolytic process for PET recycle, that has been recently patented and for which only a preliminary feasibility analysis was previously carried out, has been selected: basic data was obtained from the patent application and from open literature.

The initial frameworks for a synthesis procedure of solid and batch processes proposed respectively by Rossiter and Douglas (1986) and Malone (Douglas, 1988) have been considered. As far as a hierachical procedure to the process control system design is concerned, the papers by Fisher *et al.* (1985) and Ponton and Laing (1993) were taken into account.

The PET recycling process

Three major methods are generally adopted for PET recycling: methanolysis or hydrolysis to DMP or TPA, glycolysis to monomer and direct melt extrusion of densified waste. For producing top quality polymer (e.g. food grade), only the first and second processes can be employed: in the first case the polymer has to be reduced to the original substances, while in the second case 2-idroxyethyl terephtalate (BHET) is obtained, which allows to skip the first step of the polymerisation process.

Glycolysis to the monomer is a less demanding and less costly process than methanolysis or hydrolysis but is more versatile than direct melt extrusion (Gintis, 1992). The limits to its application for production of top quality polymer are due to high impurity content in the BHET produced by the processes proposed up to now. A modification of the glycolytic recycle process, with a more efficient purification step, was recently proposed (Pilati *et al.*, 1995): a preliminary profitability study was also carried out (Ardia, 1994; Mazzarino and Bava Pilone, 1996). A complete basic engineering study is presented in this paper, by tackling the problem according to the block diagram proposed by the previous authors, but with a different selection of the steps that need to be operated batch or continuously; attention has been focused on energy integration and on the operating procedure.

Conceptual design

Starting from the literature information and the laboratory outputs it was decided to operate batchwise with a pilot plant fed with 50 kg of PET per batch. Reasonable heat and process stream recovery were set as mandatory to demonstrate feasible operations. Minimisation of process by-products (such as the EG sludge from the evaporator) were pursued: according to such specification, a stream of the so-called "process water" (polluted to some extent by EG) circulate in the process to precipitate the BHET or to re-dissolve it without aqueous stream release from the process, though minimising the load to environment.

A conceptual cut between the batch and continuous operation was given at the interception between production (glycolysis, oligomer precipitation, BHET crystallisation and re-crystallisation, BHET discharge) and material separation and recovery (evaporation and distillation) on an accumulation of process fluid (EG-water solution): see the block diagram in Figure 1.

A given series of batch operations (up to seven/24 hours) was conceived and made possible by the overall process structure, to provide batches of BHET according to various process policies.

Basic engineering design

According to the input-output specification given in the conceptual design, the process was developed by means of the unit operations presented in the following simplified item list:

Reactor: stirred tank provided of a heating jacket to suspend and treat by glycolysis the PET scraps: batch operation with an increasing viscosity at 190°C and N_{js}=25 rpm for a mixture residence time τ=3.5 hours.

Strainer separator: 500 μm, 0.4 m^2 screen to retain fine metal and PE, PET, PVC impurities: on-line separation with the reacted mixture discharge.

Cooling tank: thermal conditioning of the EG strained mixture for the subsequent dilution with recycled process water: batch operation from 170 to about 90°C in τ=0.3 h step; the cooling medium is process water at 40°C.

Precipitator: dilutes the reacted EG with process water at the 1:3 ratio to precipitate and digest the unreacted oligomer: τ=0.5 h at 80°C.

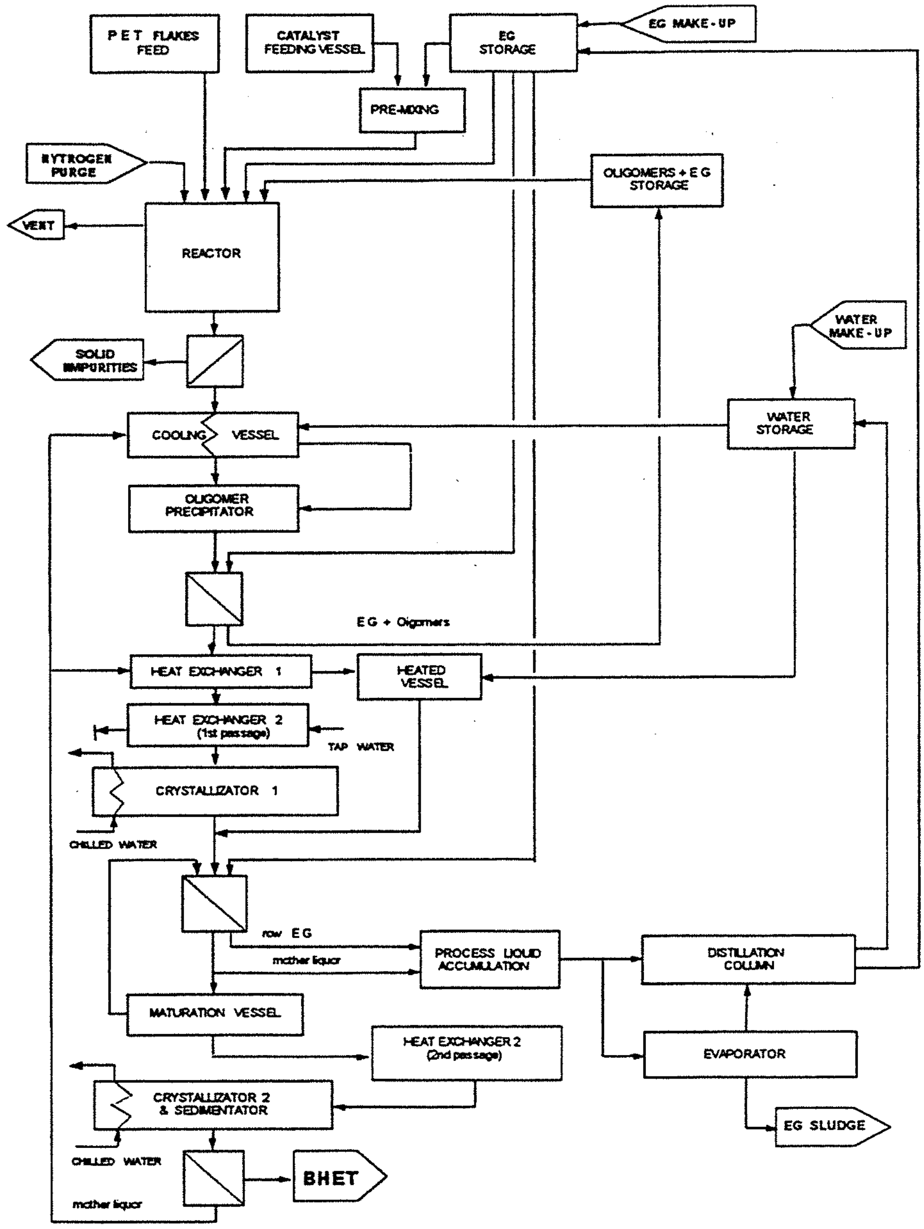

Figure 1 - Block diagram.

Disk filter: high efficiency oligomer separation to be subsequently dissolved by EG at 80°C according to a given policy.

Heat exchangers: single pass shell and tube heat exchanger for partial thermal recovery from water-EG process fluid and economy of gelid water to the downstream crystallizer.

Crystallizer 1 and 2: stirred tank for obtaining raw (#1) or refined (#2) BHET by cooling to 10°C the water-EG mixture: τ=3 h.

Filter press separation: acts on the BHET/water-EG suspension to form a 15 mm thick cake and subsequent BHET by dissolution with hot process water (>95°C) without manual operation on the solid products: $\tau_{filtration}$=0.15 h and $\tau_{dissolution}$=2 h on a 6 m^2 cloth area. The water-EG solution is stocked in a tank to feed the distillation column. EG is recovered from the mother liquor retained in the cake by an intermediate rinse with the cold water from the downstream final product separator. The filter requires an occasional hot EG treatment to dissolve oligomer accumulation.

Maturation tank: a 0.5 m^3 tank were to receive and hold for a minimum time the hot water-BHET solution to promote a thorough BHET fines dissolution.
Product separator: gravity filter to drain out the mother liquor, which is stored as process waste.
Evaporator: concentrates a fraction of the solution to be distilled in order to discharge a sludge of metallic and polymeric impurities in EG. The vapour phase is directed to the column to recover the enthalpy of the vapour phase.
Distillation column: vacuum (30 kPa), 21 tray separator, 300 mm I.D., 6 m high continuously operated up to exhausting the water-EG solution stored in the tank (3 out of 7 days/week).

P&ID details and operating book

The interaction between the process designer and the control designer brought to several modifications of the process scheme up to obtaining the conclusive outline. Each apparatus was provided of the necessary control loops and of the safety equipments; a PLC structure was devised as necessary to give the right operation sequence, because part of the process runs as batch operations.

A short operating book was compiled to illustrate the process steps and timing, as well as the logic of possible options.

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